Novel magnetic phases in multi-orbital Mott insulators

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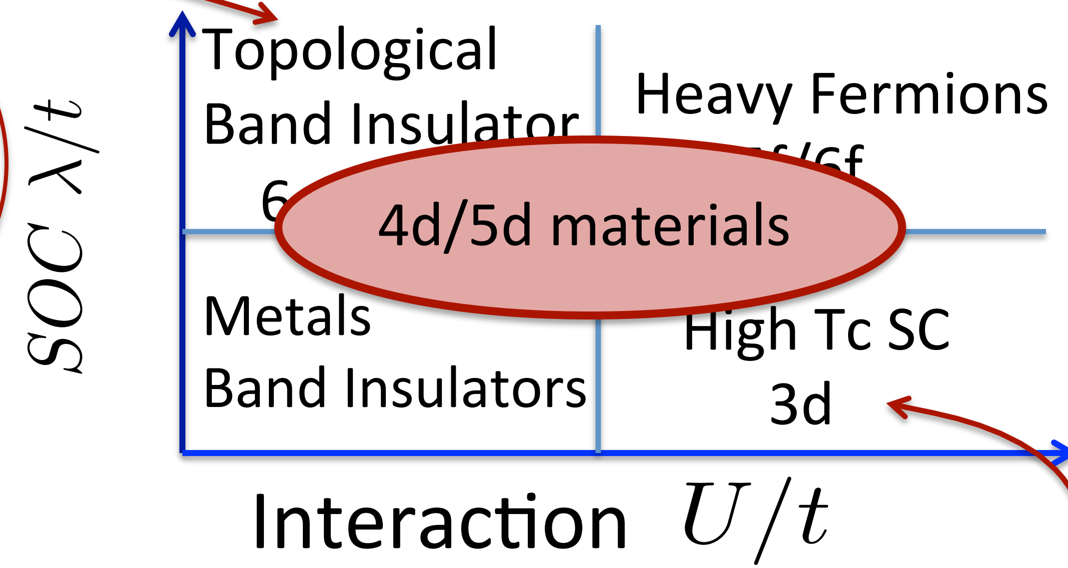
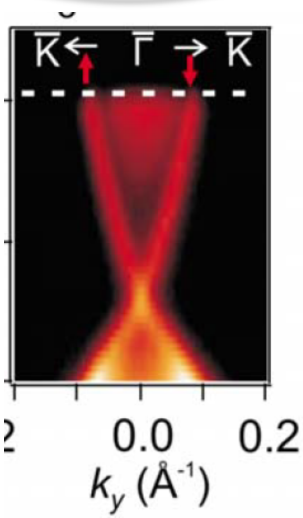
NSF MRSEC – DMR 0820414

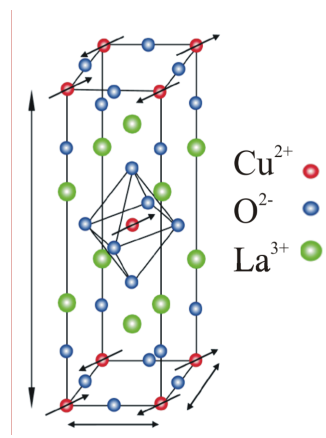
Interplay of SOC + U

New paradigm:
Topological Invariant

- Bulk band gap
- Time reversal symmetry
- Protected gapless surface states







3 broken paradigms

- band theory
- BCS theory
- Fermi Liquid theory

PART I:
d^3-d^3

Sr_2CrOsO_6 :
the highest T_c =720K
among all perovskites
with a net M.

Theory of High Tc Ferrimagnetism in a Multiorbital Mott Insulator
Meetei, Erten, Randeria, Trivedi, Woodward
PRL 110, 087203 (2013)

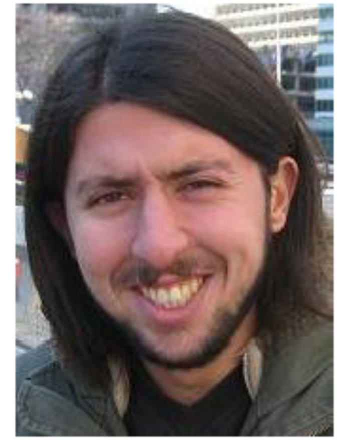
PART II
d^4

Prediction of novel orbitally entangled ferromagnetism in d^4 systems
Meetei, Cole, Randeria, Trivedi, arXiv:1311.2823

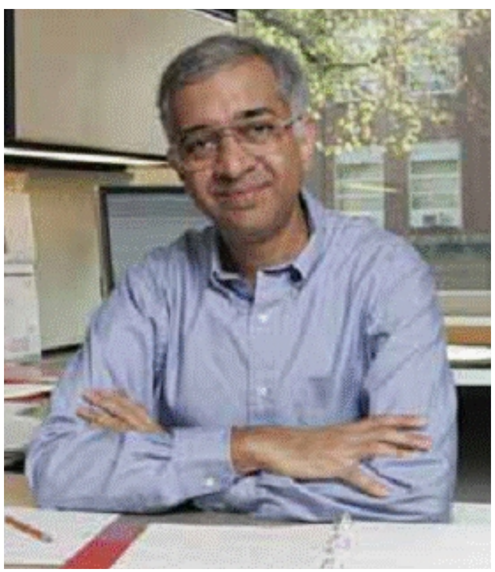
Poster

O. Nganba Meetei

William Cole



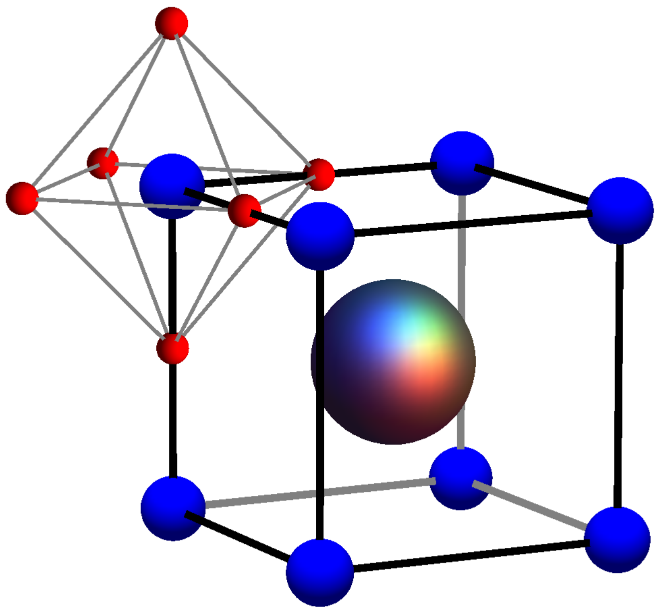
Onur Erten
(now postdoc Rutgers)



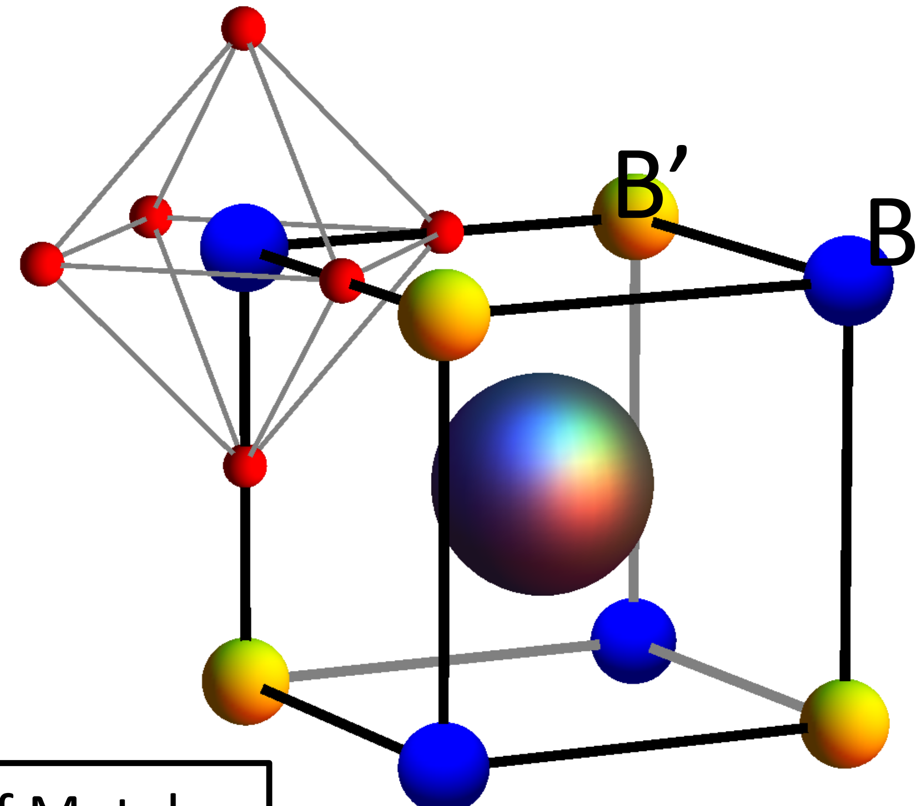
Mohit Randeria

Patrick Woodward

ABO_3 Perovskite



$(ABO_3)\ (AB'O_3)$ Double perovskite



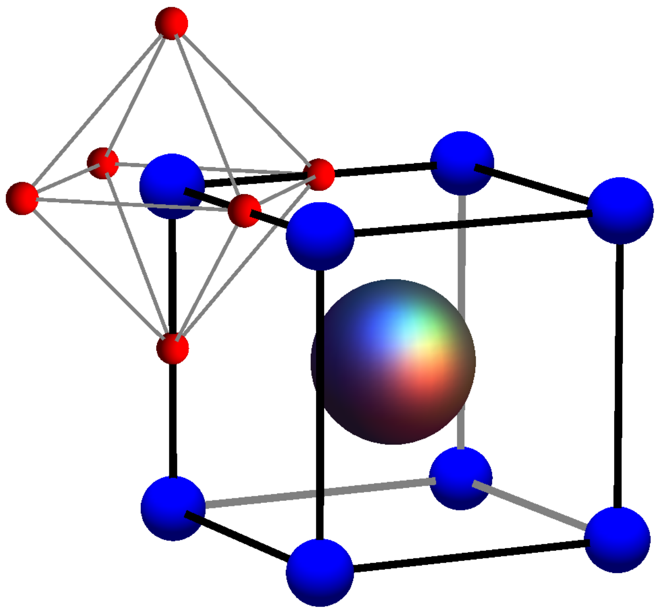
U

Cr	Mn	Fe	Co
Mo	Tc	Ru	Rh
W	Re	Os	Ir

Half Metal
Ferromagnet
Tc=420K

Sr_2FeMoO_6

ABO_3 Perovskite



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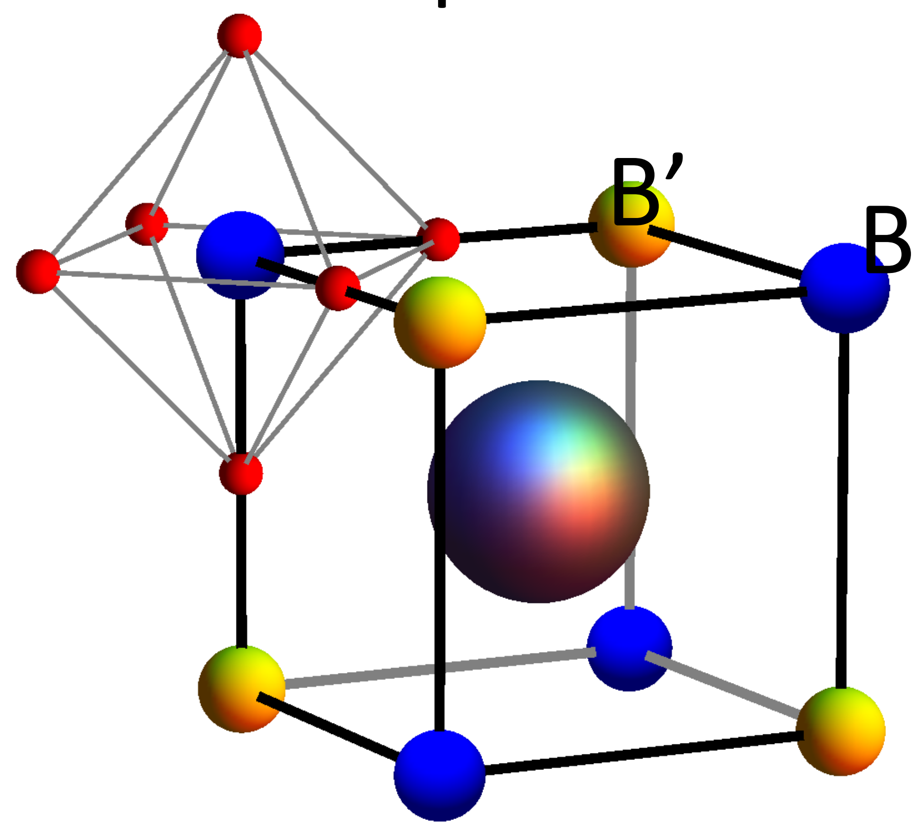
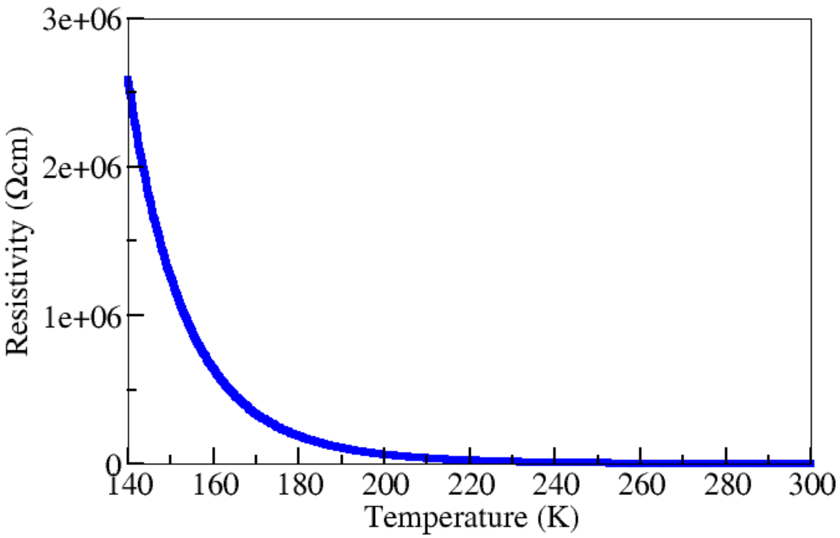


U

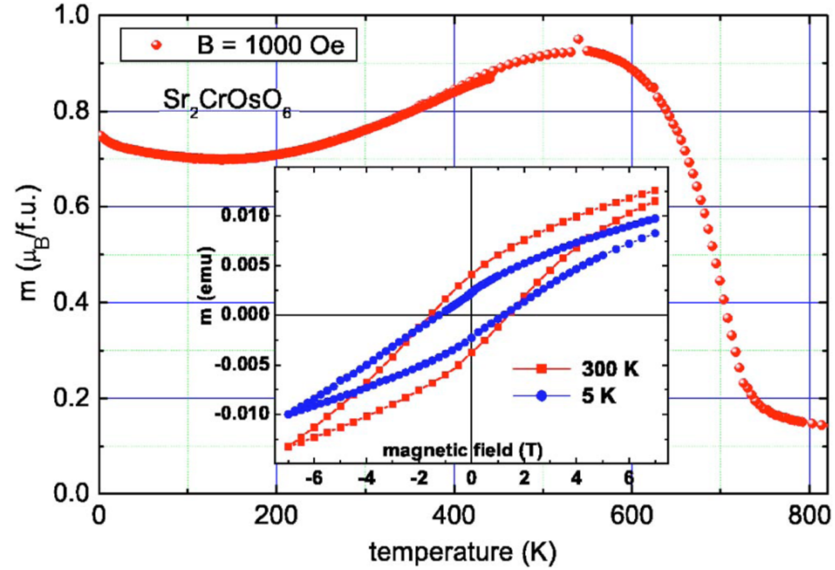
Cr	Mn	Fe	Co
Mo	Tc	Ru	Rh
W	Re	Os	Ir

$\boxed{Sr_2CrOsO_6}$

Sr_2CrOsO_6 (SCOO) puzzles



Soliz, Woodward (unpublished)



Krockenberger *et al.*
PRB **75** 020404 (2007)

Cr: $3d^3$
Os: $5d^3$ → Half filled in t_{2g}

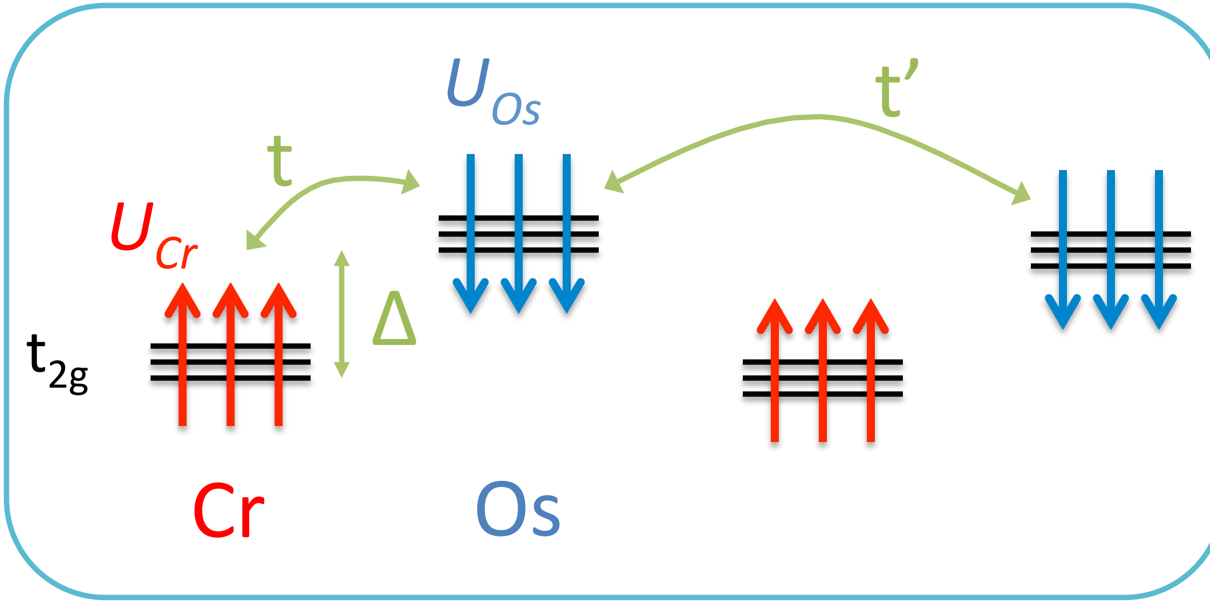
- Why is it an insulator?

- Why is there a net moment in a d^3-d^3 material? Role of spin-orbit ?

- Why is T_c = 720K so high?

- Why is M(T) non-monotonic?

Hamiltonian:



$\widetilde{U}_{Cr} = U_{Cr} + 2J^{H}_{Cr}$ = 7 eV

Os

$\widetilde{U}_{Os} = U_{Os} + 2J^{H}_{Os} - 3\lambda_{Os}$ = 2.25 eV

W = 8t = 1.5 eV

U Values taken from:
Imada, Fujimori, Tokura RMP 1998;
Lee & Pickett EPL 2007

Importance of J_H at half filling:
de' Medici, Mravlje, Georges, PRL 2011

Hierarchy of Scales

- Charge sector: Effective *U*'s on Cr & Os
  → new Mott criterion
  → SCOO is a multi-band Mott insulator

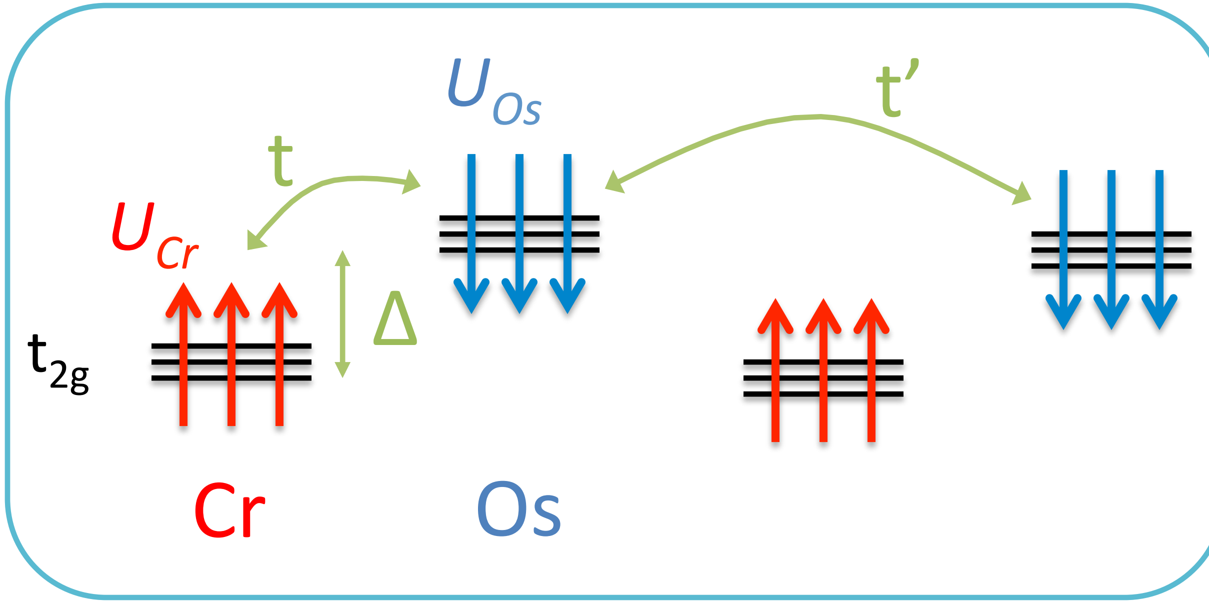
  Slave Rotor mean field theory

- Spin at a single site: Hund's coupling → S=3/2 local moments
  Spin-orbit coupling → L is quenched no L.S coupling

- Spin-spin interactions:
  Antiferromagnetic superexchange
  Frustration: Canted AF ground state
  non-monotonic M(T)

*Charge Sector: Slave-rotor mean field theory**

To treat the strong correlations:
enlarge the Hilbert space subject to a constraint



$c_{i\alpha}^{\dagger} = f_{i\alpha}^{\dagger}\, e^{i\theta_i}$ → *rotors*
→ *spinons*

⇒ $H(f_{i\alpha}, \theta_i), \quad L_i = \sum_{\alpha} f_{i\alpha}^{\dagger} f_{i\alpha}$ constraint

treat constraint within mean field:

$$H \simeq H_S(f_i, \langle\cos(\theta)\rangle) + H_R(\cos(\theta_i), \langle f^{\dagger} f\rangle)$$

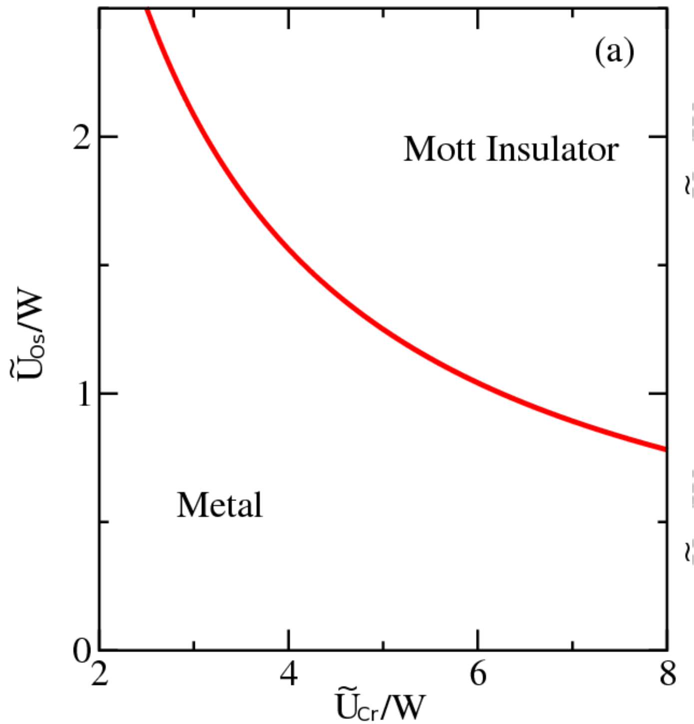
Coupled self-consistently

$\langle\cos(\theta)\rangle \neq 0$ Metal

$\langle\cos(\theta)\rangle = 0$ Insulator

*For standard Hubbard model: S. Florens & A. Georges PRB **70** 035114 (2004)

Charge Sector: New Mott Criterion



Analytical Solution

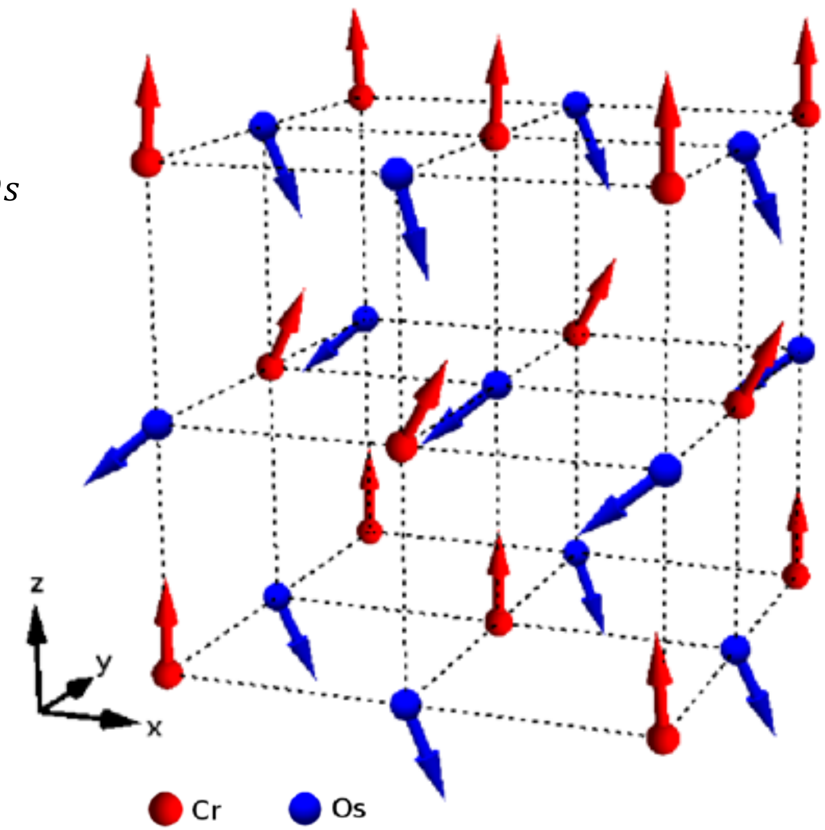
$$\sqrt{U_{Cr} \cdot U_{Os}} > \ 2.5\ W$$

W = 8t = 2D Bandwidth

- For single perovskites ⇨ $U > 2.5\ W$
- Small U on Os can be compensated with large U on Cr.
  - Sr2CrOsO6 is a Mott insulator

Frustrated Antiferromagnetic Interactions:

$$H_{eff} = J_1 \sum_{<ij>} S_i^{Cr} \cdot S_j^{Os} + J_2 \sum_{\langle\langle ij \rangle\rangle} S_i^{Os} \cdot S_j^{Os}$$



- J_1 & J_2 are both AF
- J_2 leads frustration

Monte Carlo (4096 Spins)
Variational calculation

Frustration ⇨ Canted Ground state
Net moment (T=0)
Extra peak in S(q)

Frustrated Antiferromagnetic Interactions:

$$H_{eff} = J_1 \sum_{<ij>} S_i^{Cr} \cdot S_j^{Os} + J_2 \sum_{\ll ij \gg} S_i^{Os} \cdot S_j^{Os}$$

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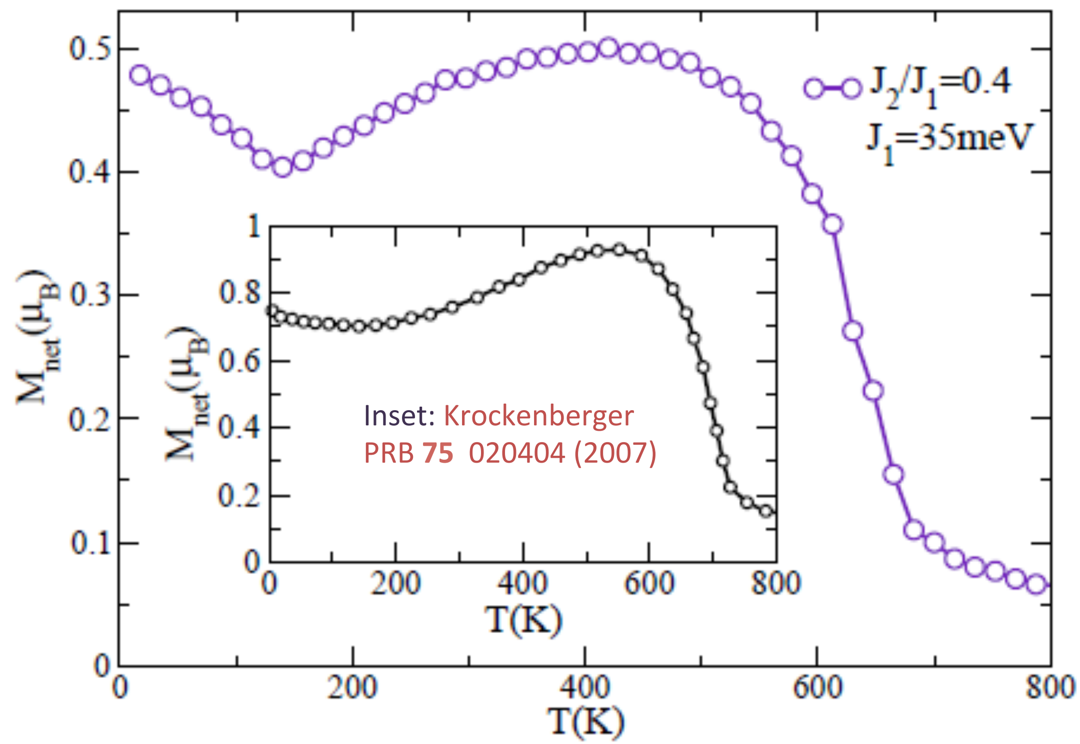


Frustration $\Rightarrow$ Canted Ground state
Net moment (T=0)
Extra peak in S(q)
Non-monotonic M(T)

Sr_2CrOsO_6 Summary

Insulator?	✓ Multi-band Mott Insulator $\sqrt{U_{Cr} \cdot U_{Os}} > \ 2.5\ W$
Net moment with d^3-d^3?	✓ Canting due to frustration - Os-Os AF superexchange
Spin-orbit?	✓ L is quenched, no SO (confirmed by XMCD)
High T_c?	✓ intermediate U
Non-monotonic M(T)?	✓ Frustration

Proposal for neutron diffraction:
A new extra peak in S(q): (π, π, 0)/a due to canting

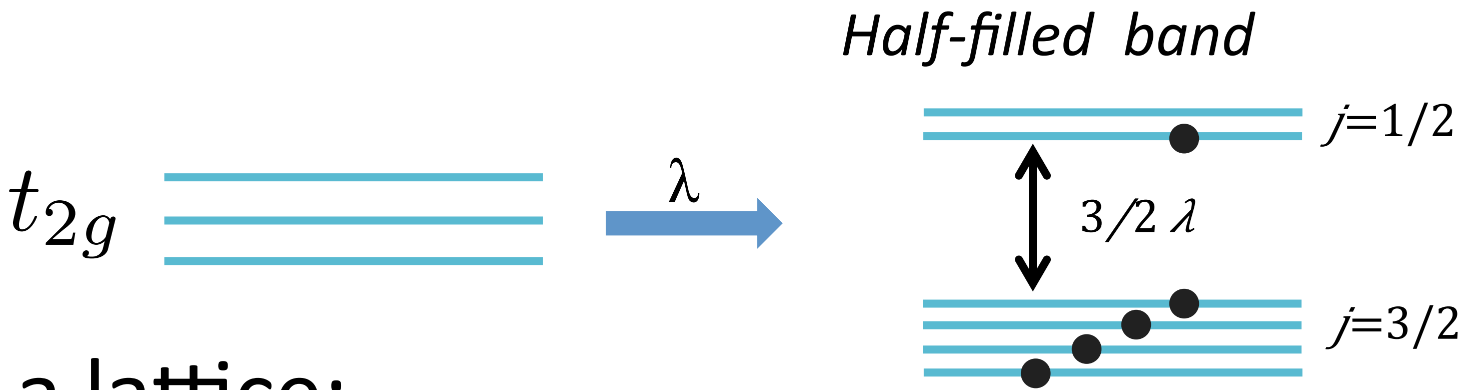
PRL 110, 087203 (2013)

PART II

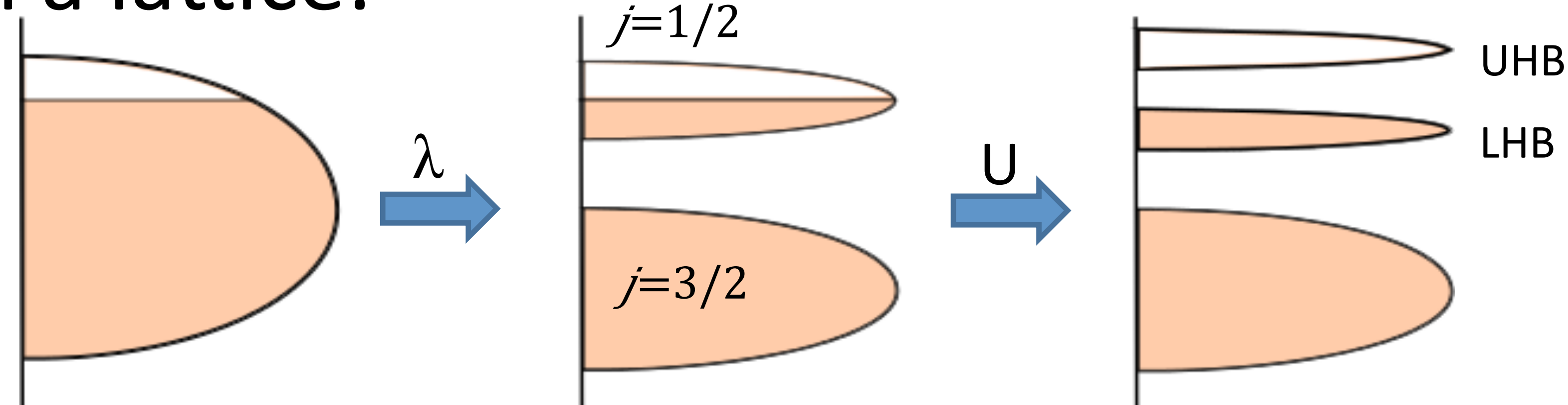
d^4

SOC + U in d^5 (iridates)

Atomic Limit:



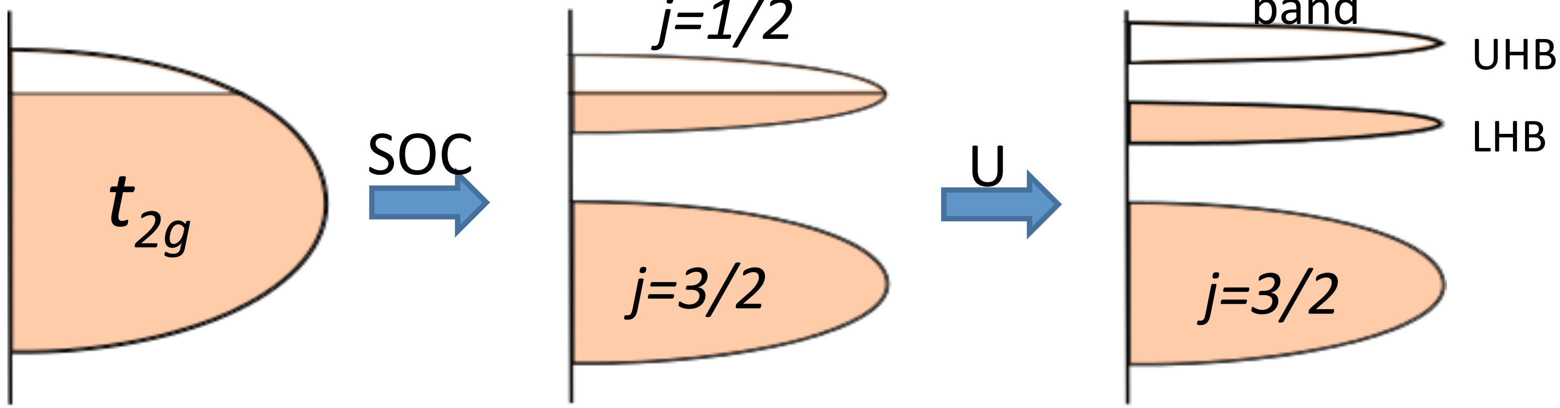
On a lattice:



SOC assisted Mott insulator
e.g. Sr_2IrO_4

S_2IrO_4 ($5d^5$)

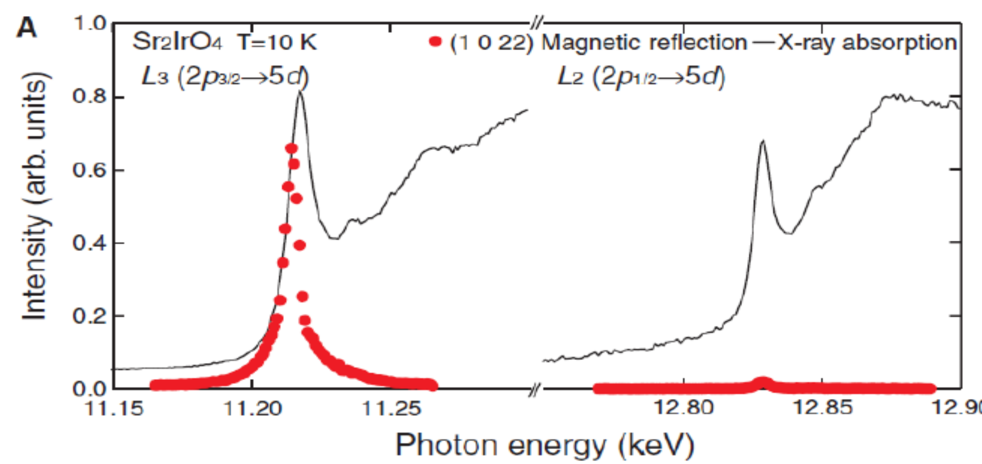
Moderate U opens gap in SOC split band



Resonant X-ray Scattering (RXS)

Uniquely identify j=1/2 state

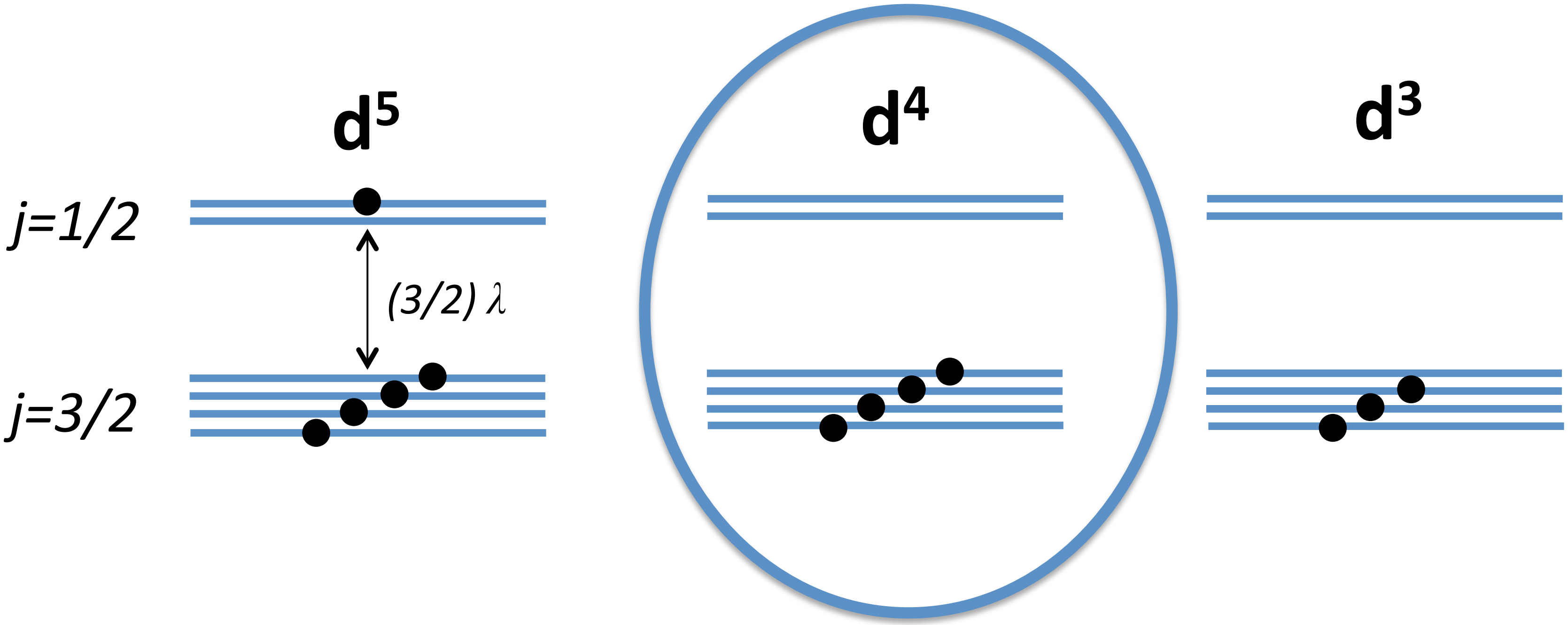
Completely suppressed L_2 edge



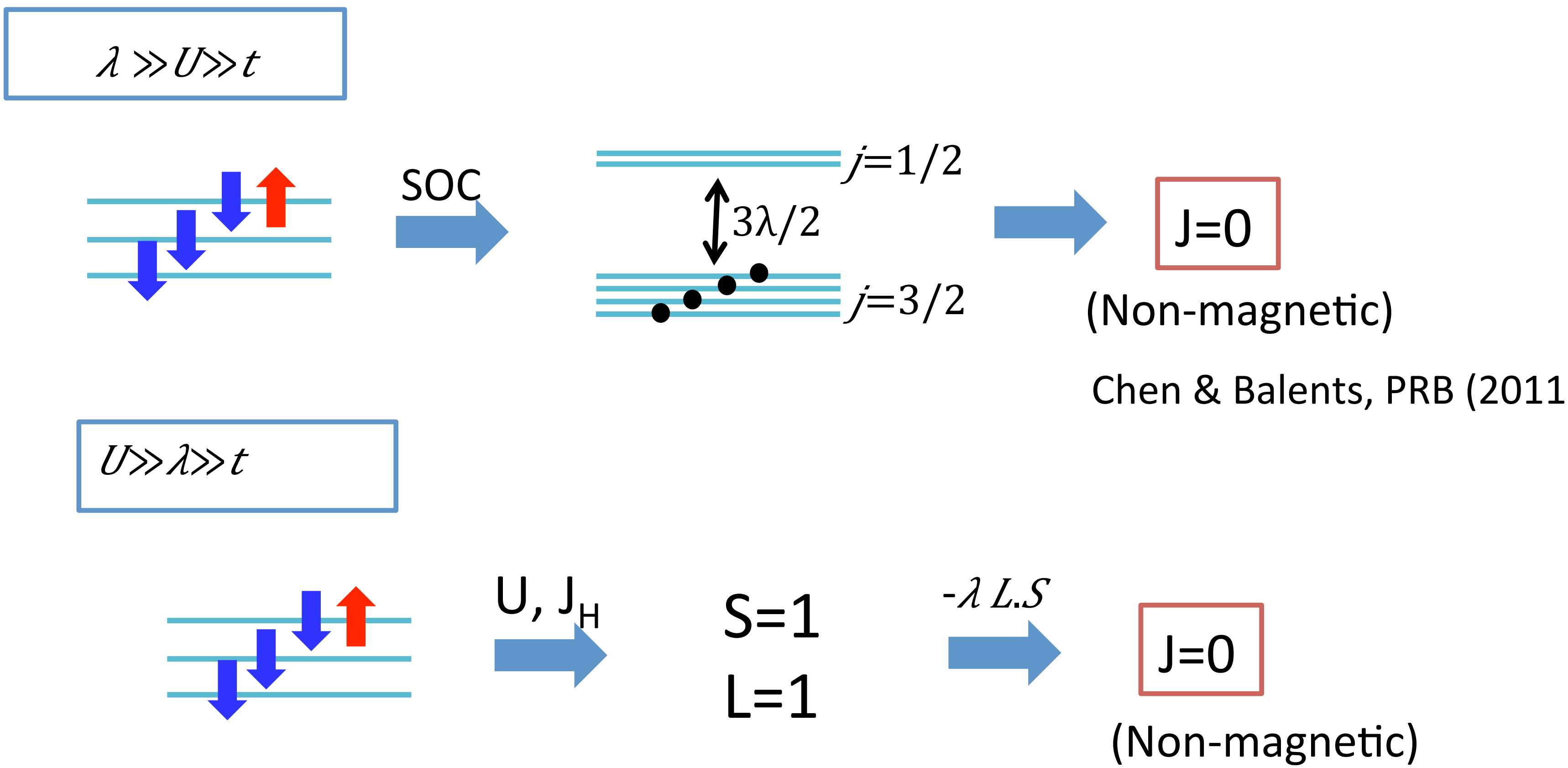
Kim, B. J. *et al.*, *Science* (2009)

Kim *et al.* Science **323**, 1329 (2009)

What about other fillings?



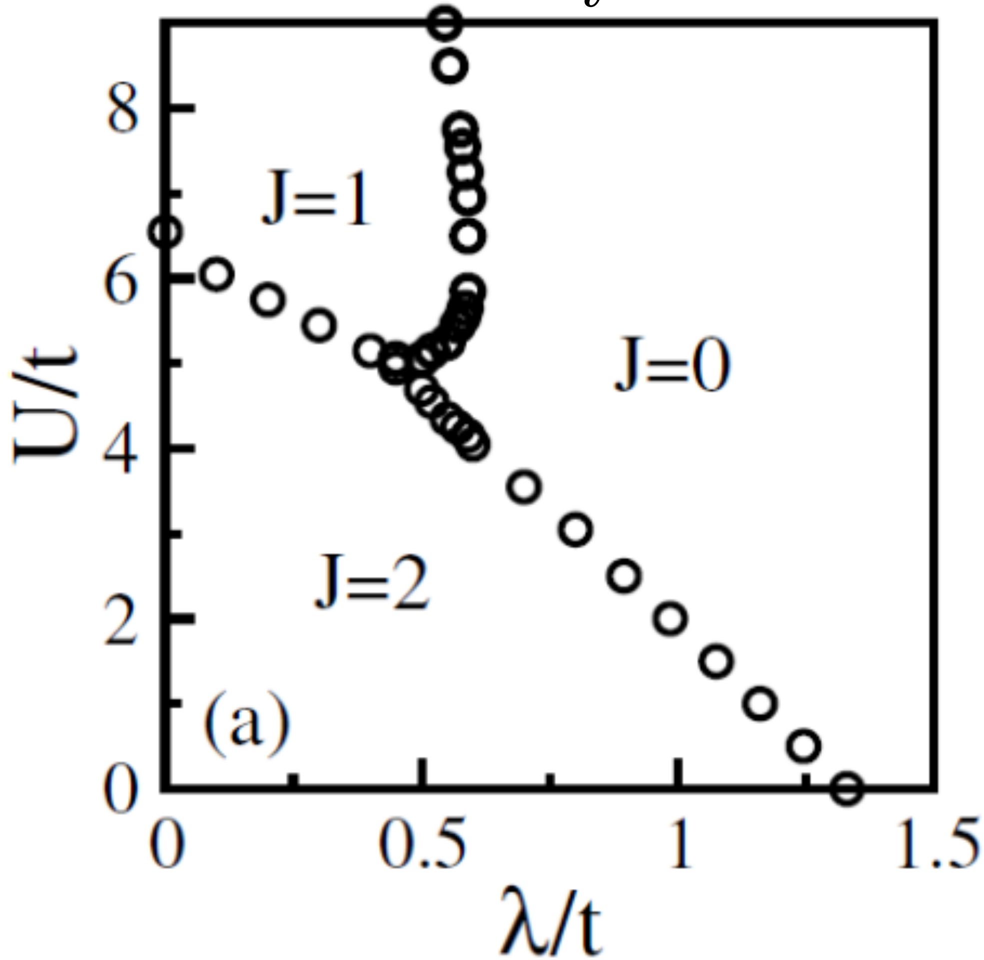
d^4 systems are non-magnetic in atomic limit



Can there be any non-trivial magnetism in d^4 systems?

Hopping induced Ferromagnetism in d4 system

$$H = H_{hop} + \sum_i (H_{i,U} + H_{i,SOC})$$



d^4: 2 sites

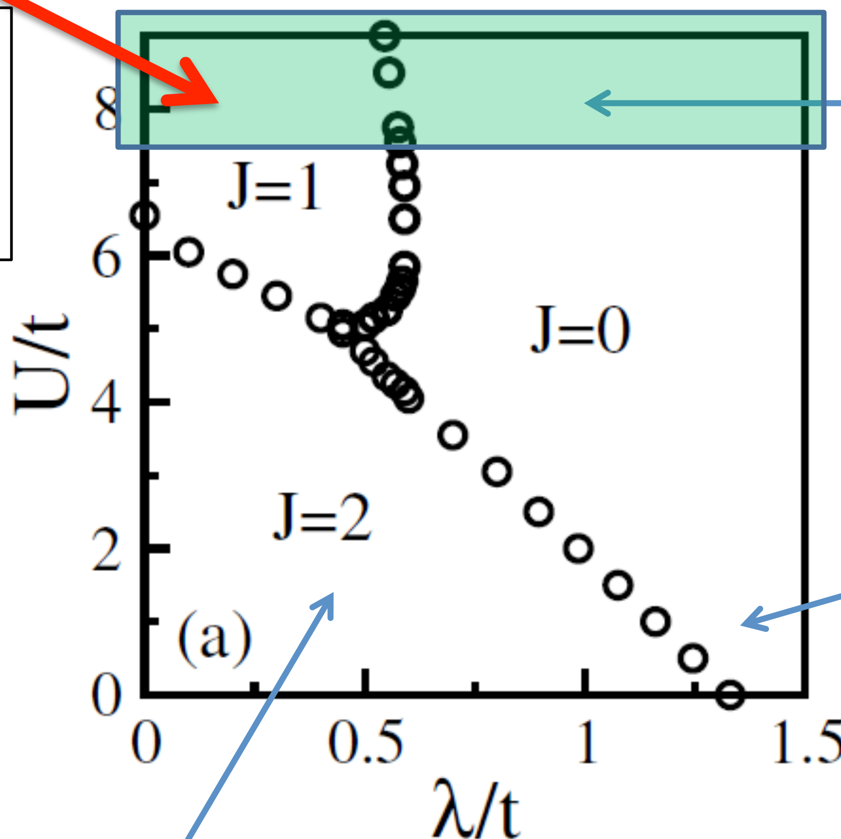
Exact diagonalization

$${}^{(6\times 2)}C_8 \approx 500$$

states

Hopping induced magnetism

NOVEL FM



Atomic picture expected to work

SOC Driven Band Insulator ($CaOsO_3$)
Band theory

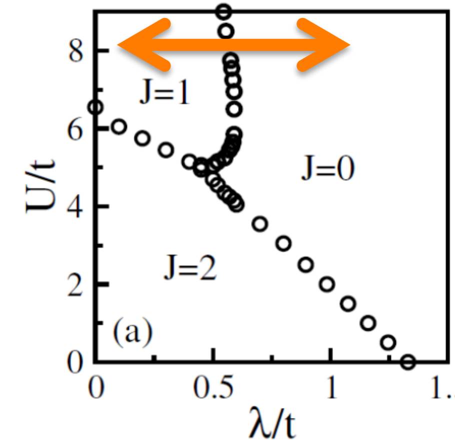
Stoner Ferromagnet ($SrRuO_3$)
Mean field theory

Meetei, Cole, Randeria, Trivedi, arXiv:1311.2823

Why novel FM?

(1) Hopping generates a local moment

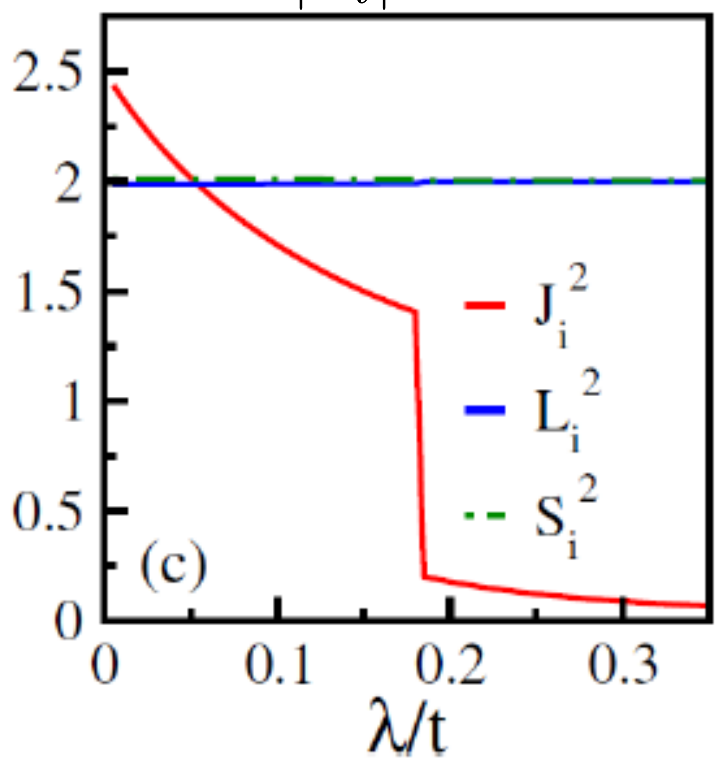
(2) Local moment is not robust



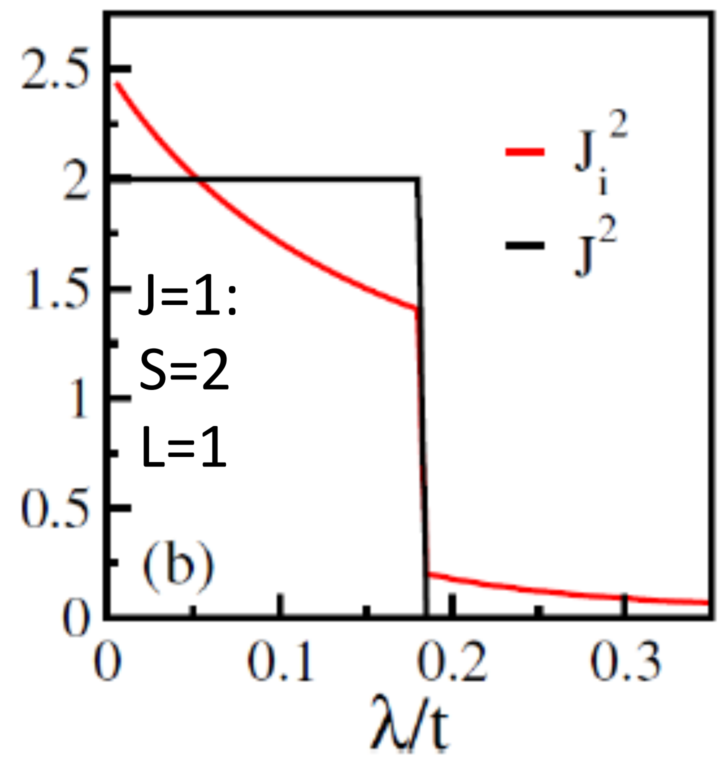
U/t=8

$$|L_i| = 1$$

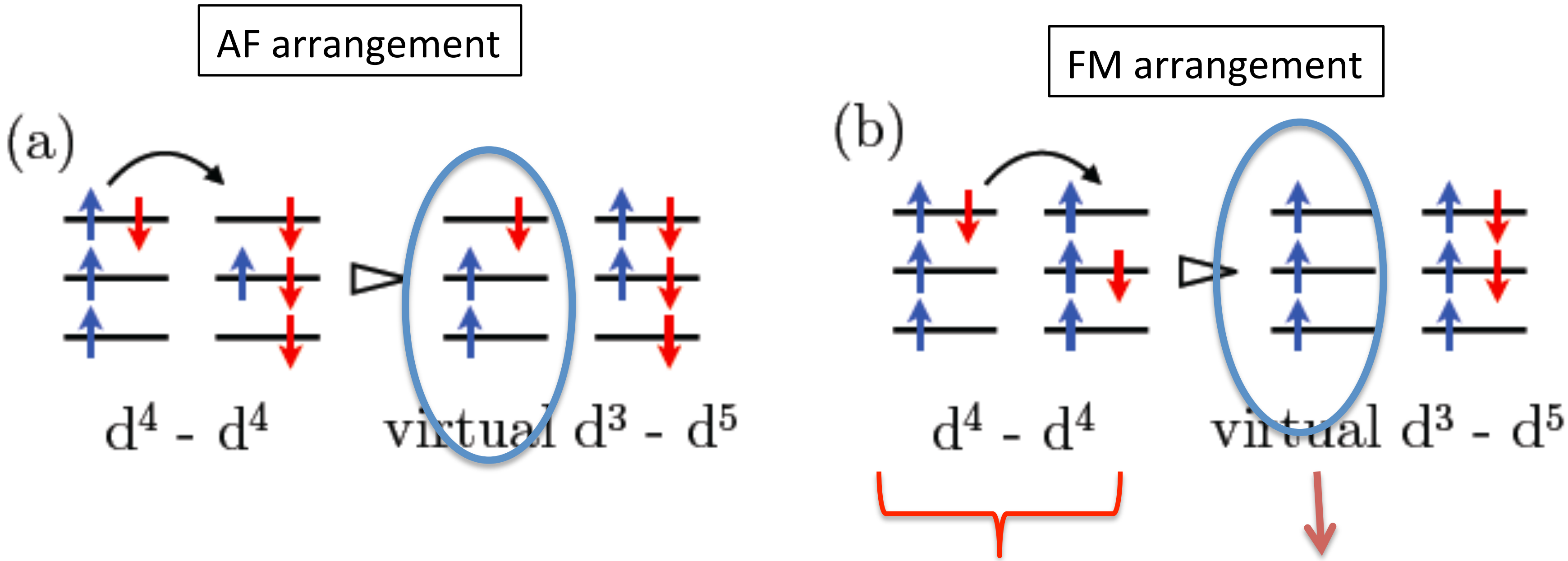
$$|S_i| = 1$$



(3) Local moments are coupled ferromagnetically



Why *ferromagnetic* superexchange?

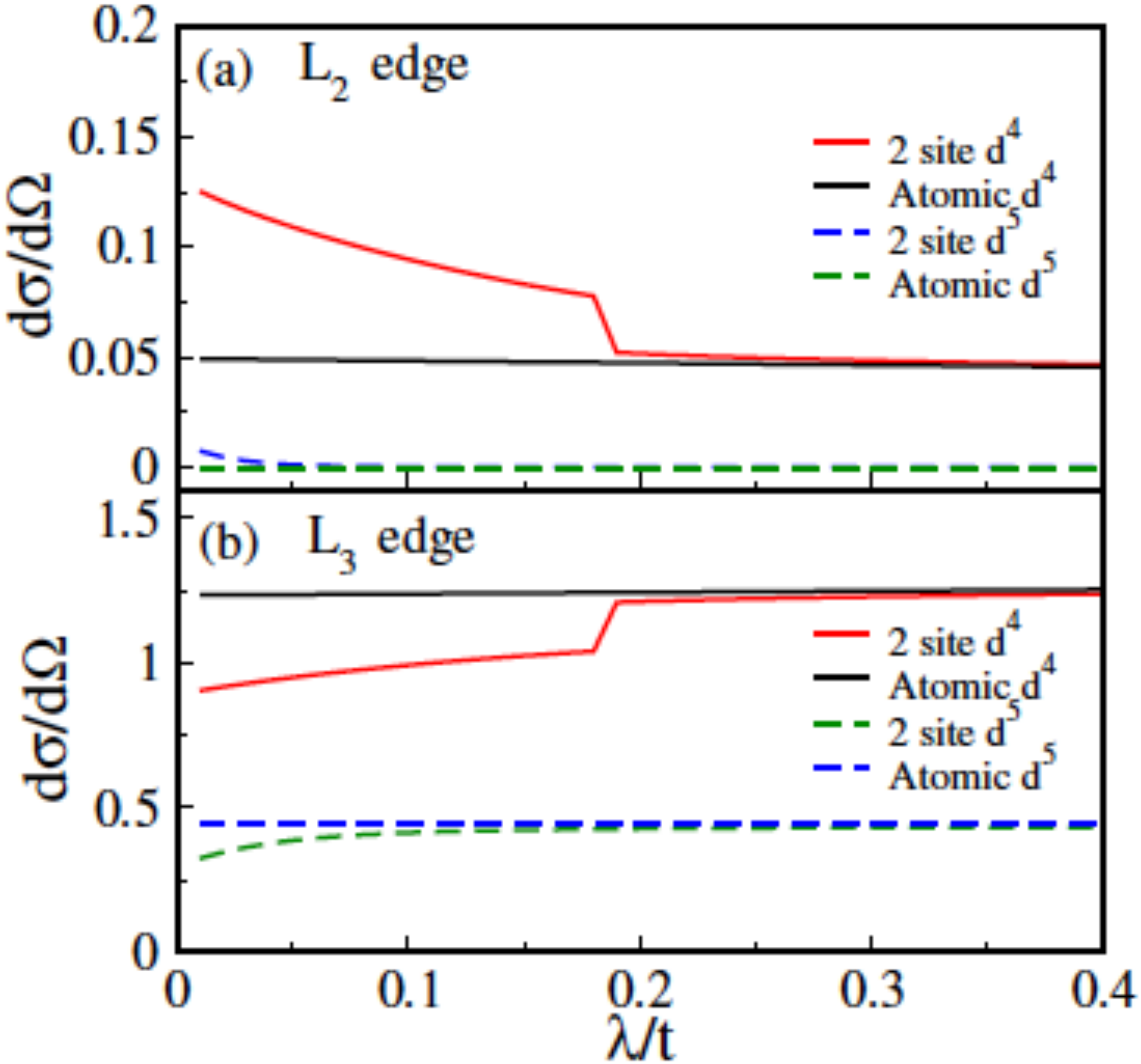


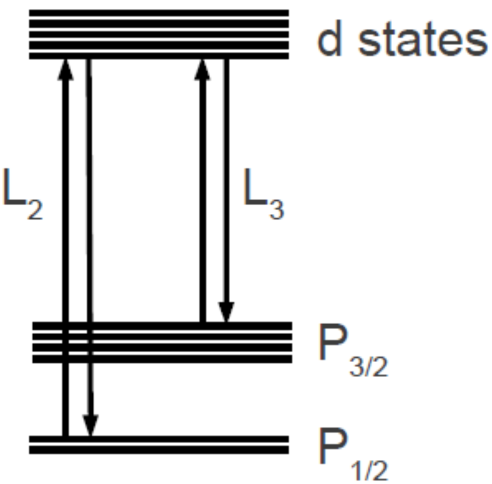
For maximum gain the two electrons on nbr sites must be in different orbitals

Hund's coupling MAXIMIZED

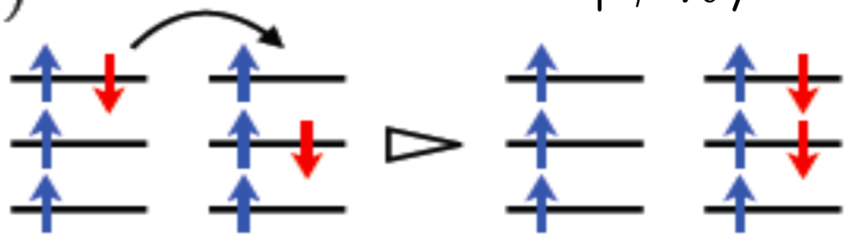
In contrast G. Khaliullin PRL 111, 197201 (2013) claims AF superexchange

RXS spectra: Atomic vs 2 Site d4 vs d5





b) GS: $|\psi_n\rangle$



d^4 - d^4 virtual d^3 - d^5

$L_i = 1$ $L_i = 1$ $L_i = 0$ $L_i = 1$

$S_i = 1$ $S_i = 1$ $S_i = 3/2$ $S_i = 1/2$

$$\tilde{H}' = H_{hop}\left[\sum_n \frac{|\psi_n\rangle\langle\psi_n|}{E_G - E_n}\right] H_{hop}$$

$$\boxed{\tilde{H} \approx -J_{FM}\mathbf{S_1}\cdot\mathbf{S_2}\mathcal{P}(\mathbf{L_1}+\mathbf{L_2}=1)}$$

Orbitally entangled Ferromagnet

S=2 antialigned with L=1

Effective Hamiltonian for d^4 system

Superexchange + SOC

$$H_{eff} = -\frac{J_{FM}}{2}\sum_{\langle ij\rangle}\mathbf{S}_i\cdot\mathbf{S}_j\mathcal{P}(\mathbf{L}_i+\mathbf{L}_j=1)+\frac{\lambda}{2}\sum_i\mathbf{L}_i\cdot\mathbf{S}_i$$

Competition between SOC and J_{FM}:

SOC		**FM superexchange**
$L_1 + S_1 = 0$ and $L_2 + S_2 = 0$	competes with	$S_1 + S_2 = 2$ and $L_1 + L_2 = 1$

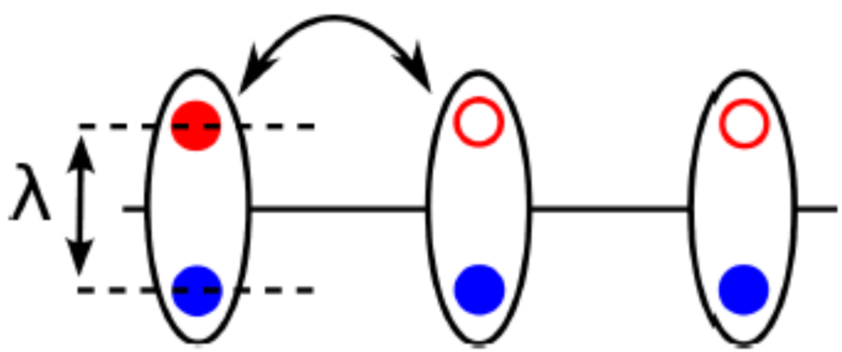
Competition between SOC and J_{FM} → Magnetic phase transition

Mean-field theory for effective Hamiltonian

Locally L=1 + S=1 → J=0, 1 and 2 (Ignore high energy J=2)

$$S_i^\alpha = -\sqrt{\frac{2}{3}}\left(T_{i\alpha}^\dagger s_i + s_i^\dagger T_{i\alpha}\right) - \frac{i}{2}\epsilon_{\alpha\beta\gamma} T_{i\beta}^\dagger T_{i\gamma}$$
$$L_i^\alpha = \sqrt{\frac{2}{3}}\left(T_{i\alpha}^\dagger s_i + s_i^\dagger T_{i\alpha}\right) - \frac{i}{2}\epsilon_{\alpha\beta\gamma} T_{i\beta}^\dagger T_{i\gamma}$$

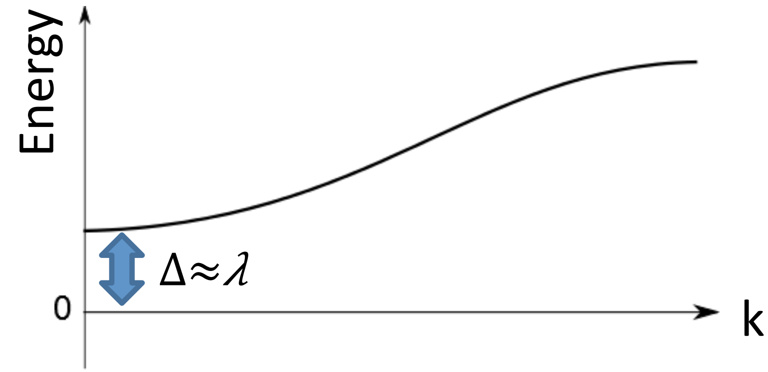
α = x,y,z

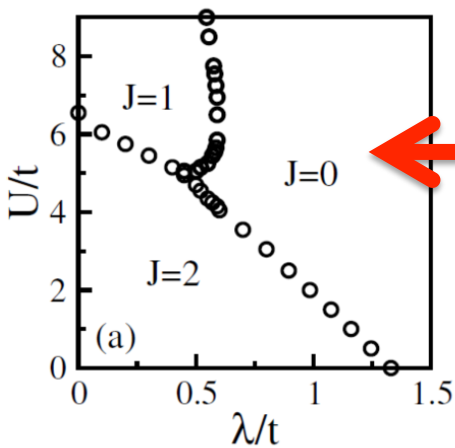


Sachdev, Bhatt PRB 41, 9323 (1989)

Mean-field theory for effective Hamiltonian

$\boxed{(J_{FM}/\lambda) < (J_{FM}/\lambda)_C}$





- Singlet condensate
- Gapped triplet band

Mean-field theory for effective Hamiltonian

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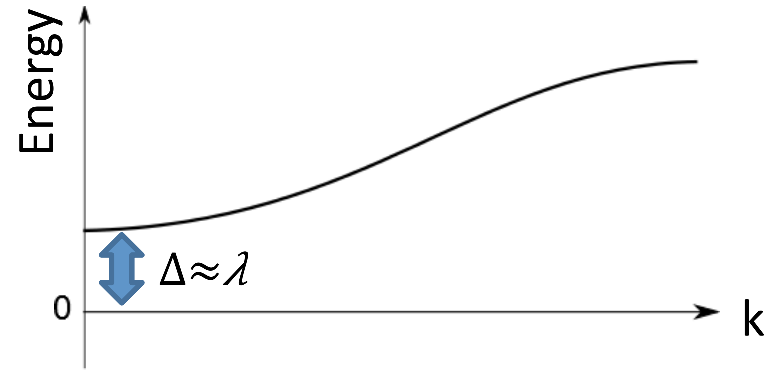
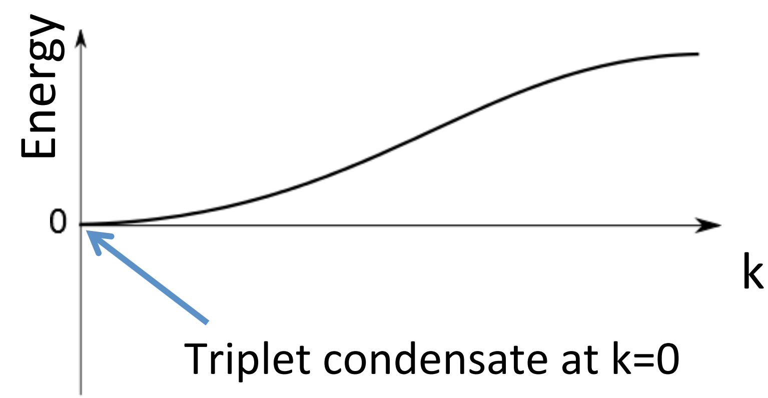


$\boxed{(J_{FM}/\lambda) = (J_{FM}/\lambda)_C}$

- Triplet gap closes
- Triplet condensate forms

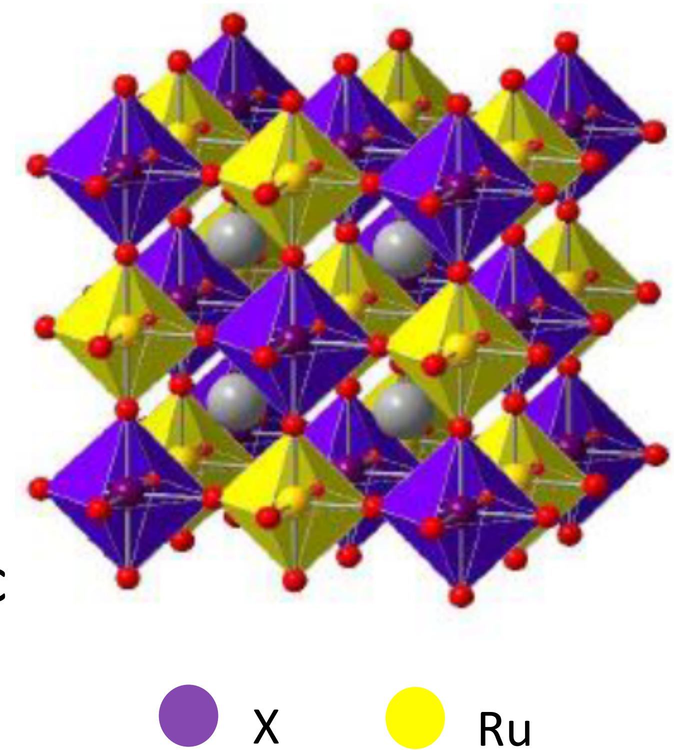


See also G. Khaliullin PRL 111, 197201 (2013)

Candidate materials for d^4 ferromagnetic Mott insulators

La_2XRuO_6 where X=Zn,Mg

- Ru in d^4 configuration
- X ion is in d^0 state → suppressed hopping
- Mott insulator
- La_2ZnRuO_6 is close to ferromagnetic transition. Both ferromagnetic and nonmagnetic samples exist.



R. I. Dass *et al.* Phys. Rev. B (2004).
K. Yoshii *et al.* Phys. Stat. Sol.(a) (2006).

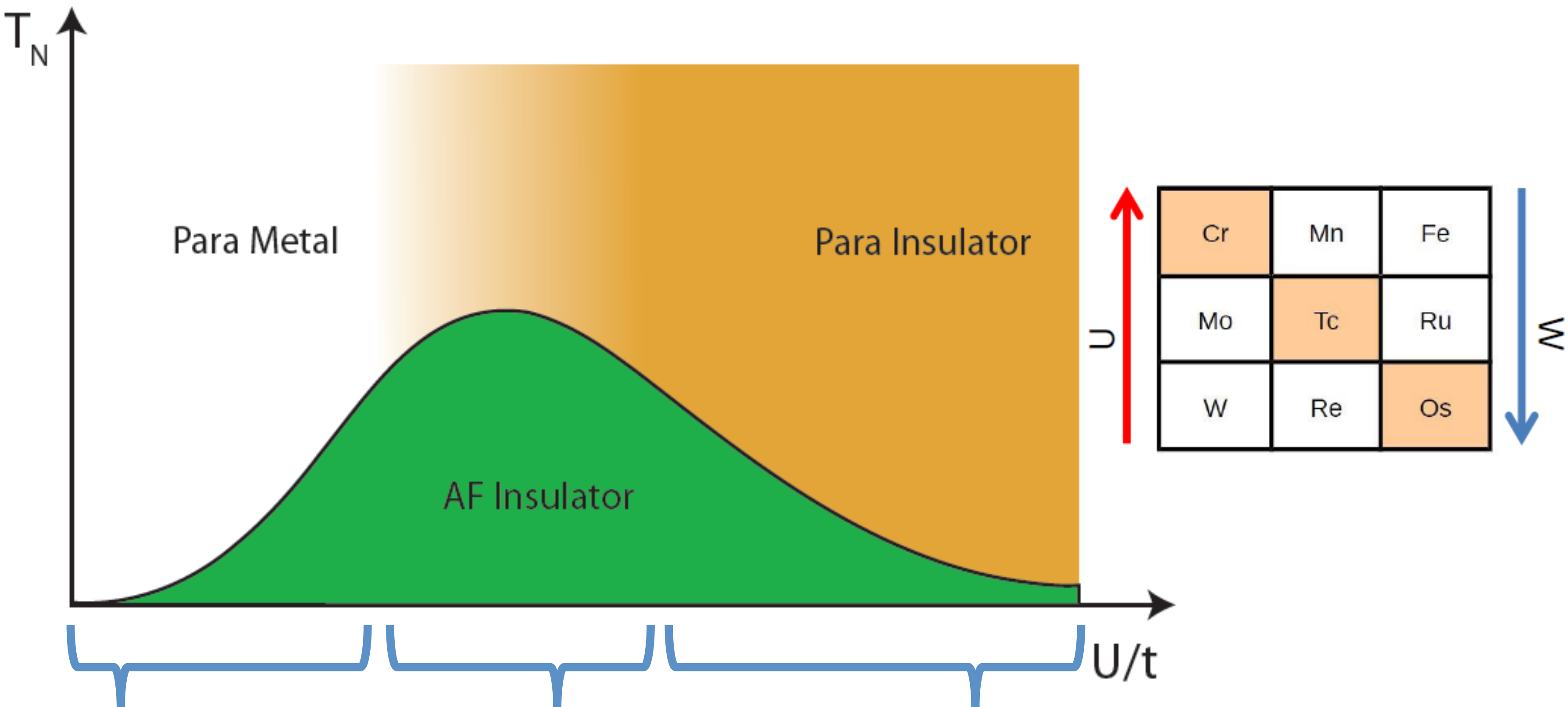
Novel Magnetism in d^4 Mott insulators

Summary

- Orbitally entangled ferromagnetism in d^4 Mott insulator
- Kinetic energy stabilizes local moment in contrast to non-magnetic atomic limit.
- Effective Hamiltonian for d^4 Mott insulators.
- Predictions for unique RXS signatures
- Candidate materials

Meetei, *et al.* arXiv:1311.2823 (2013)

Stuff….



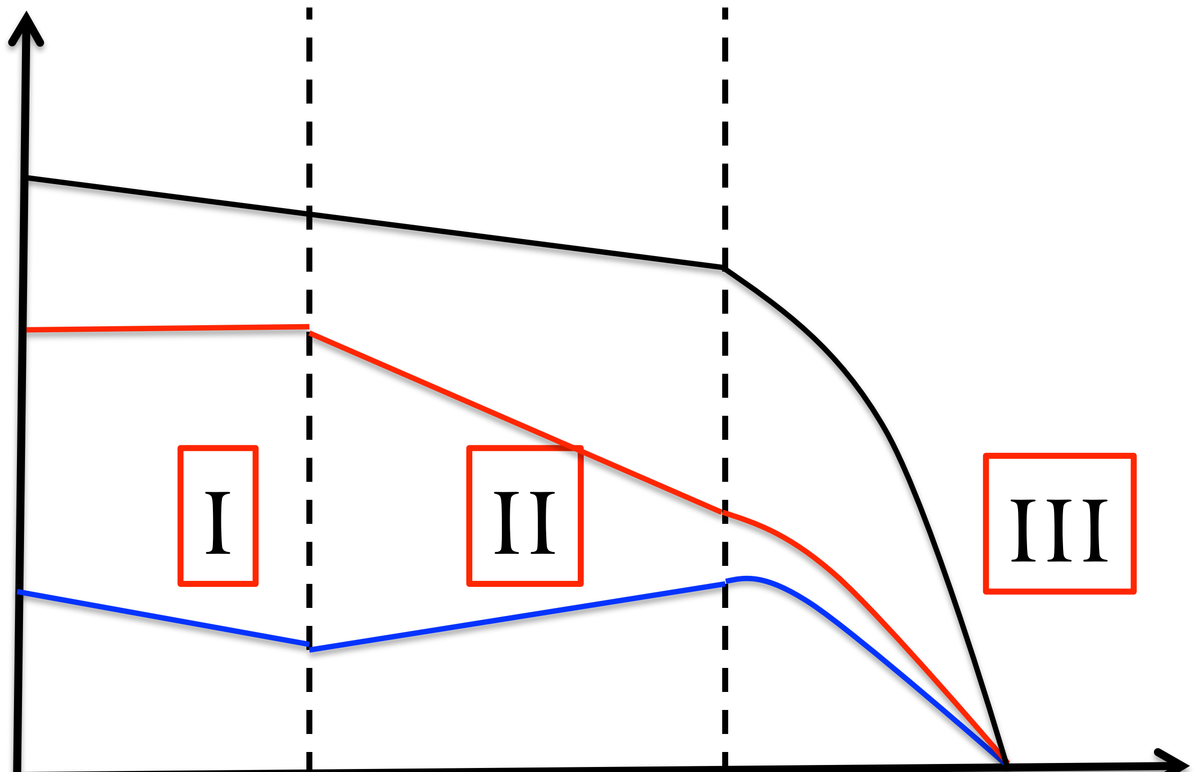
Weak coupling (Slater)
$NaOsO_3$ ($5d^3$): T_N=410K

Intermediate coupling (W~U)
$SrTcO_3$ ($4d^3$): T_N=1000K
Sr_2CrOsO_6($3d^3$-$5d^3$): T_C=720K
$U_{eff}=(U_{Cr}U_{Os})^{1/2}$

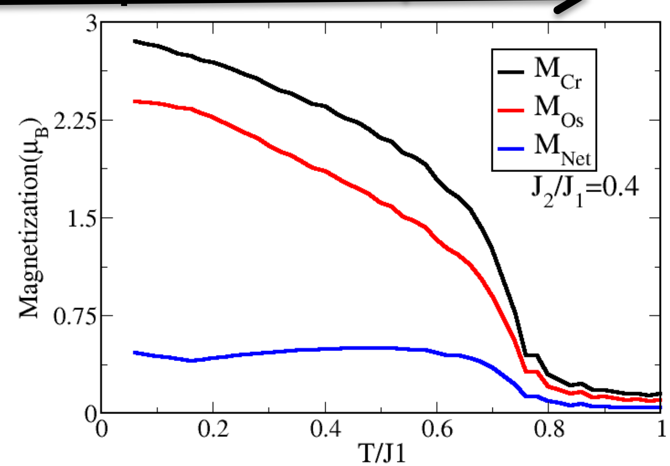
Strong coupling (Mott)
$LaCrO_3$($3d^3$): T_N=320K

Combining 3d-5d tunes U_{eff} to intermediate U where T_C is largest!

Non-monotonic M(T)



$M_{net} = M_{Cr} - M_{Os}$



I - Low T

Os sublattice gets stuck in canted state, while Cr keeps on ordering.

II - Intermediate T

Spin stiffness:

$J_{Cr} \sim J_1$ & $J_{Os} \sim J_1 - J_2$

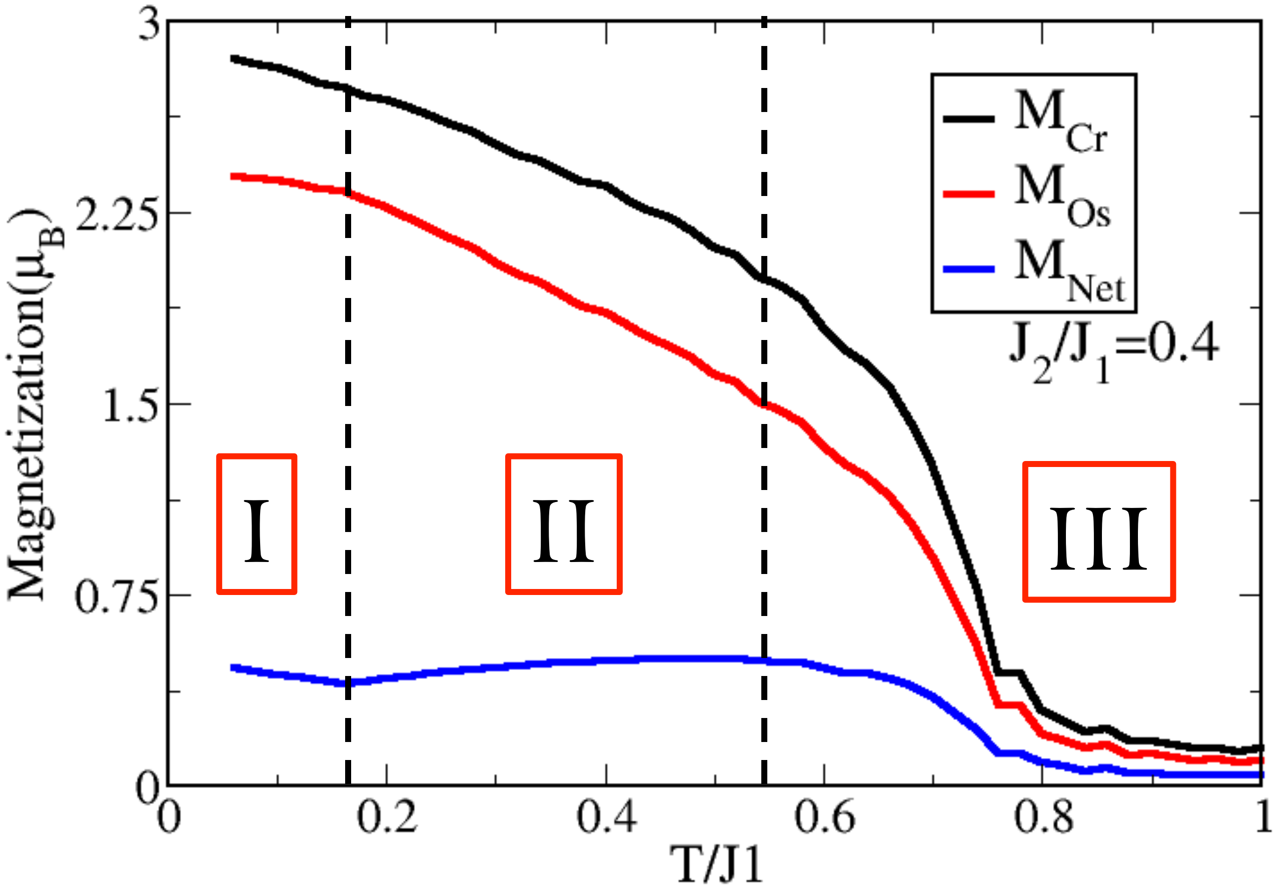
Os spins are floppy

$M_{net} = M_{Cr} - M_{Os}$ increases

III - High T

Both Cr & Os drops rapidly close to T_c.

Non-monotonic M(T)



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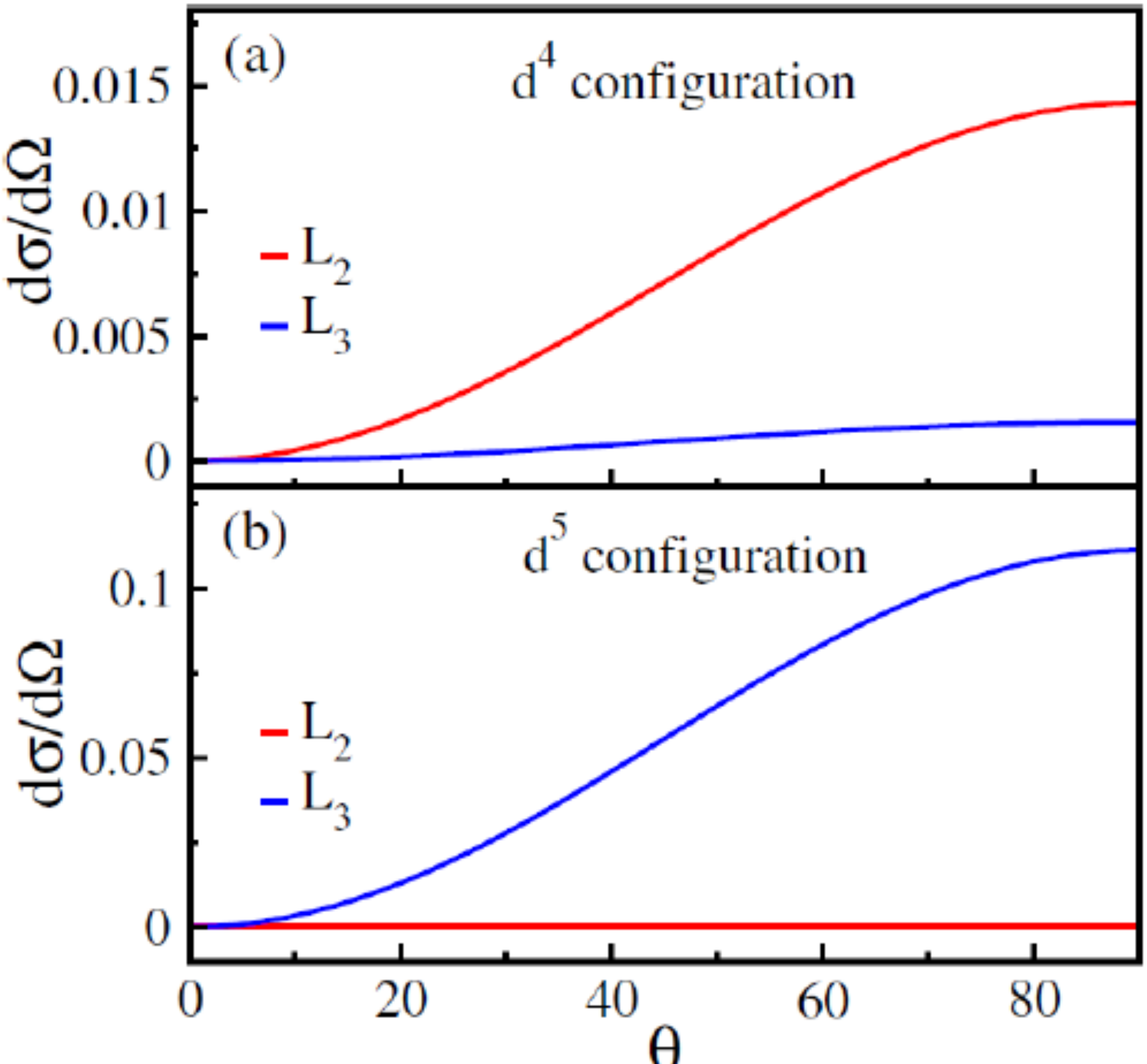
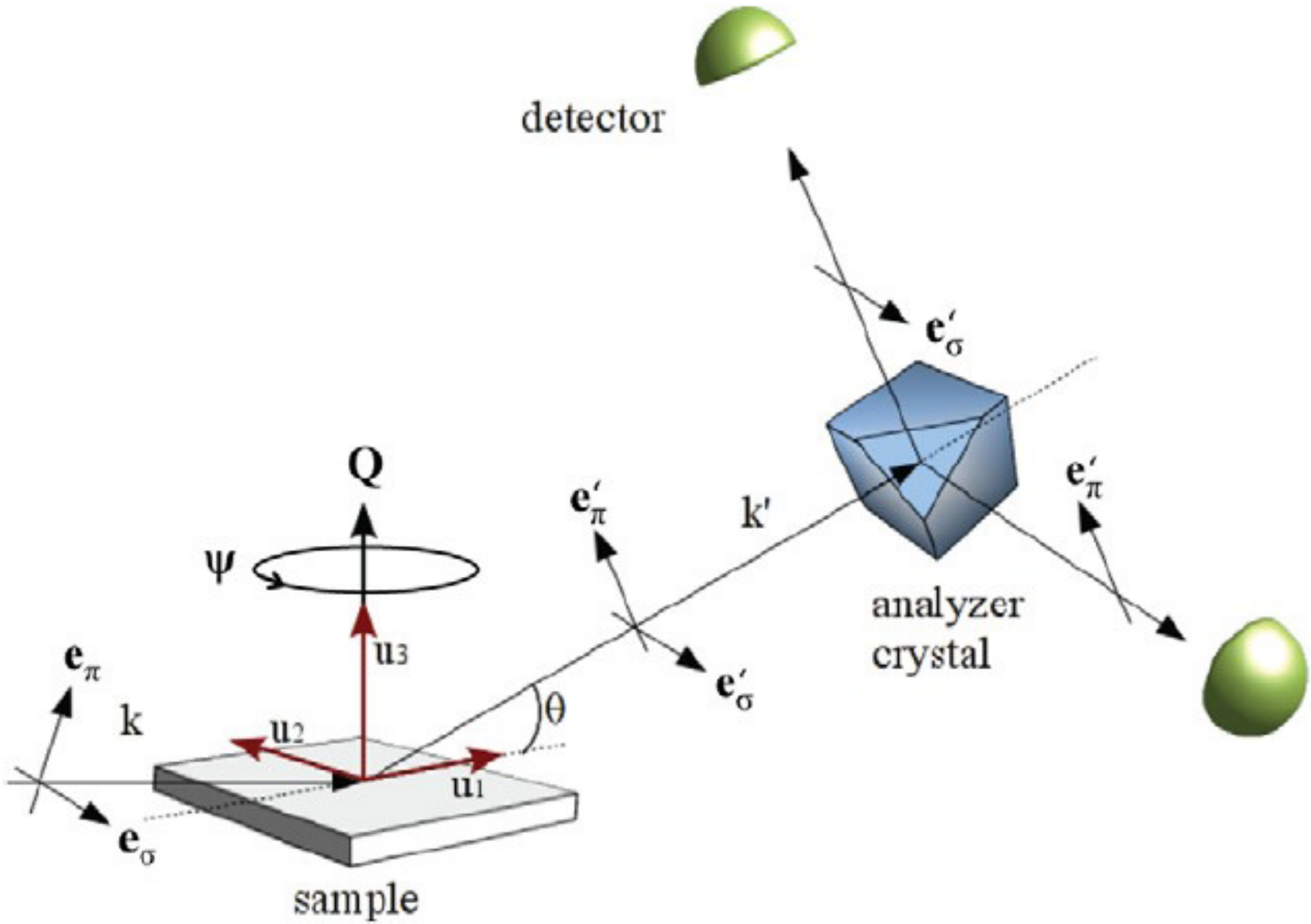
Magnetic RXS $(\pi \rightarrow \sigma)$



Scattering geometry



Resonant enhancement

Free ion approximation (usually good for Mott insulators)

$$\Delta f(\omega) \propto \sum_n \frac{\langle \Psi_G | (\mathbf{e}'.\mathbf{D})^\dagger | \psi_n \rangle \langle \psi_n | \mathbf{e}.\mathbf{D} | \Psi_G \rangle}{E_n - E_G - \hbar\omega - i\Gamma}$$

When effect of neighboring sites are strong

$$\Delta f(\omega) \propto Tr \left[\rho \sum_n \frac{(\mathbf{e}'.\mathbf{D})^\dagger | \psi_n \rangle \langle \psi_n | \mathbf{e}.\mathbf{D}}{E_n - E_G - \hbar\omega - i\Gamma} \right]$$